Multiphoton Process Observed in the Interaction of Microwave Fields with the Tunneling between Superconductor Films

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Calculations are made which explain qualitatively the multiphoton-assisted electron tunneling recently observed in superconducting diodes by Dayem and Martin. It seems to us that the microwave field is much too weak to cause any nonlinearities in the conduction current in the superconductors. Thus, the interaction does not cause transitions between electron states with different wave numbers. Rather, the energies of the electrons are varied adiabatically by the microwave fields. This gives rise to effective changes in the density of states versus energy which are dramatically illustrated in the tunneling current.

Calculations are performed for three different possible forms of the field interaction. Qualitatively, the theory fits the experimental observations very well, but, as in the somewhat similar case of phonon-assisted tunneling, the largest postulated interaction seems about an order of magnitude too small to explain the observations on a quantitative basis.

I. INTRODUCTION

IN a recent Letter,[1] Dayem and Martin reported experiments on the tunneling between superconducting films in the presence of microwave fields, and show evidences of absorption or emission of one or more photons by a single tunneling electron. The phenomenon resembles one observed earlier in silicon Esaki junctions,[2] where indirect tunneling transitions through a combination of transverse acoustic and optical phonons have been detected. A similar multiphonon effect in the tunneling current has also been reported in the superconducting $Al\text{-}Al_2O_3\text{-}Pb$ sandwiches by Rowell, Chynoweth, and Phillips.[3] In this paper we focus our attention to the multiphoton process observed in the superconducting diodes.

Dayem and Martin used diodes similar to those reported by Giaever[4] and also by Nicol, Shapiro, and Smith.[5] A typical diode is illustrated in Fig. 1. Two superconducting films, A and B, each about 100 Å thick, were insulated from one another by a somewhat thinner layer of aluminum oxide. The diode was placed inside a microwave cavity and microwave power was fed into it. Experiments were carried out at three different frequencies: 24.2, 33.4, and 55 kMc/sec, respectively. The photon energy, i.e., $\hbar\omega$, is smaller than the energy gap of either superconductor at all three frequencies. The tunneling current between the two superconductors is observed as a dc voltage, applied across the diode, is varied slowly. The results obtained in a typical case are reproduced in Fig. 1. The dashed and the solid traces shown in the figure are, respectively, the tunneling current versus the applied voltage observed with and without the microwave field.

In the presence of a microwave field, the following facts must be explained: (1) an excess of tunneling current in the region below the knee of the curve (see Fig. 1) and a reduction of the tunneling current in the region above it, and (2) the tunneling current appears in voltage steps of $(\hbar\omega/e)$, where ω is the angular frequency of the microwave field and e is the electronic charge. In the theory which follows, we assume that the fields are reasonably uniform in the region of interest, and that the field merely modulates adiabatically the energy levels of the electrons. In recent experiments a variety of field distributions in the vicinity of the diode were investigated by Dayem and Martin.[6] In order to study possible effects of different field distributions we shall consider the following three idealized cases: (1) The predominant microwave field is an electric field normal to the conducting surfaces of the diode, (2) there is an electric field parallel to the conducting surfaces of the diode, and (3) a propagating microwave field travels inside the diode, which acts as a strip line. In all the

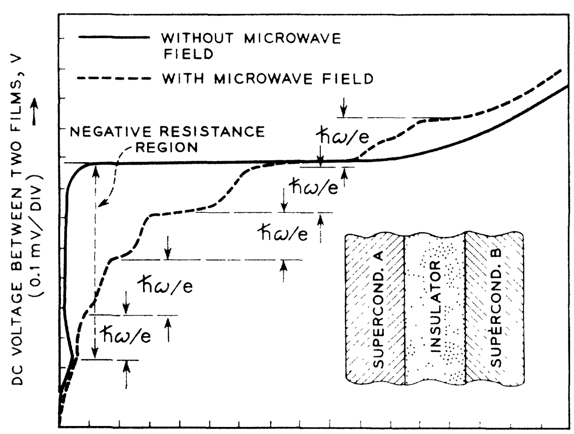

FIG. 1. Bias voltage vs tunneling current of a superconducting $Al\text{-}Al_2O_3\text{-}In$ diode as measured by Dayem and Martin with and without the microwave field. $\hbar\omega/e=0.16$ mV.

[1] A. H. Dayem and R. J. Martin, Phys. Rev. Letters **8**, 246 (1962).
[2] A. G. Chynoweth, R. L. Logan, and D. E. Thomas, Phys. Rev. **125**, 877 (1962).
[3] J. M. Rowell, A. G. Chynoweth, and J. C. Phillips, Phys. Rev. Letters **9**, 59 (1962).
[4] I. Giaever, Phys. Rev. Letters **5**, 147, 464 (1960).
[5] J. Nicol, S. Shapiro, and P. H. Smith, Phys. Rev. Letters, **5**, 461 (1960).
[6] A. H. Dayem and R. J. Martin (unpublished).

cases, we are able to explain qualitatively all the phenomena described above. Quantitatively, however, it seems that all the effects considered here are at least an order smaller than those observed experimentally.

II. EFFECT OF AN ELECTRIC FIELD ACROSS THE DIODE (CASE I)

We start with a very simple case in which an electric field is excited between the two superconducting films normal to their surfaces. We neglect the effect of all other fields. In this case, the electric field sets up a potential difference,

$$V \cos\omega t, \tag{1}$$

between the two films. For convenience, the films are labeled by A and B, respectively.

To compute the tunneling current, we must consider wave functions of quasi-particles (or excitations) in the superconductors. The energy levels of those particles are distributed below and above an energy gap and the density of states is peaked at the two edges of the gap, To facilitate the later discussion we may simply take the semiconductor model and consider the quasi-particles as the electrons and holes of the superconductor. Since the wave functions drop off very sharply in the insulating region, the interaction between the electrons and the microwave field is quite small and may be neglected there. If we neglect this interaction and hold the potential of film A as the reference, the only effect of the microwave field is to add an electrostatic potential of the form (1) to the electrons in film B.

Consider an electronic quasi-particle of energy E in film B. Suppose that it has a wave function (without the microwave field)

$$\psi(x,y,z,t)=f(x,y,z)e^{-iEt/\hbar}, \tag{2}$$

which satisfies the unperturbed Hamiltonain H_0. In the presence of the microwave field, the Hamiltonian becomes

$$H=H_0+eV\cos\omega t. \tag{3}$$

It is obvious that the interaction Hamiltonian of the form (3) does not change the spatial distribution of the wave function. The new electronic wave function satisfying (3), therefore, has the form

$$\psi(x,y,z,t)=f(x,y,z)e^{-iEt/\hbar}\Big(\sum_{n=-\infty}^{n=+\infty} B_n e^{-in\omega t}\Big). \tag{4}$$

After substituting (4) into the Schrödinger equation

$$H\psi=i\hbar(\partial\psi/\partial t), \tag{5}$$

we find

$$2nB_n=(eV/\hbar\omega)(B_{n+1}+B_{n-1}), \tag{6}$$

or

$$B_n=J_n(eV/\hbar\omega), \tag{7}$$

where J_n is the nth order Bessel function of the first kind. The new electronic wave function for electrons in film B is, therefore, in the form

$$\psi(x,y,z,t)=f(x,y,z)e^{-iEt/\hbar}\Big[\sum_{n=-\infty}^{n=+\infty} J_n(\alpha)e^{-in\omega t}\Big], \tag{8}$$

where

$$\alpha=eV/\hbar\omega. \tag{9}$$

We see that the wave function is normalized, since

$$\Big[\sum_{n=-\infty}^{n=+\infty} J_n(\alpha)\Big]^2=J_0^2(\alpha)+2\sum_{n=1}^{\infty}J_n^2(\alpha)=1 \tag{10}$$

is independent of α.

The wave function in the presence of the microwave field contains components which have energies, E, $E\pm\hbar\omega$, $E\pm 2\hbar\omega,\cdots$, etc., respectively. Without the field, an electron of energy E in superconductor B can only tunnel to the states in superconductor A of the same energy. Now with the microwave field, the electron may tunnel to the states in film A of energies E, $E\pm\hbar\omega$, $E\pm 2\hbar\omega,\cdots$, etc. Let $\rho(E)$ be the unperturbed density of states of the superconductor B. In the presence of the microwave field, we have then an effective density of states given by

$$\rho'(E)=\sum_{n=-\infty}^{\infty}\rho(E+n\hbar\omega)J_n^2(\alpha).$$

It is interesting to note that if we replace the time-dependent part of the wave function in (2) by

$$\exp\left\{-\frac{i}{\hbar}\left[Et+\int_0^t eV\cos\omega t' dt'\right]\right\},$$

and then expand the latter in a series of Bessel functions, we obtain exactly the same result as in (8) This is to be expected. The interaction Hamiltonian does not change the spatial distribution of the wave function; it can only modify adiabatically the energies of the electrons.

III. TUNNELING CURRENT

According to the Giaever[4] experiment or using the theories by Bardeen,[7,8] and by Cohen, Falicov, and Philips,[9] the tunneling current between two superconducting films may be put in the following form:

$$I_{AB}=C\int_{-\infty}^{+\infty}[f(E-eV_0)-f(E)]\times\rho_A(E-eV_0)\rho_B(E)dE. \tag{11}$$

Here C is the proportionality constant, V_0 is the dc applied voltage between the films, the f's are the Fermi factors, and ρ_A and ρ_B are densities of states of superconductors A and B, respectively. In presence of the microwave field, using (8), it is easy to show that the

[7] J. Bardeen, Phys. Rev. Letters **6**, 57 (1961).
[8] J. Bardeen, Phys. Rev. Letters **9**, 147 (1962).
[9] M. H. Cohen, L. M. Falicov, and J. C. Phillips, Phys. Rev. Letters **8**, 316 (1962).

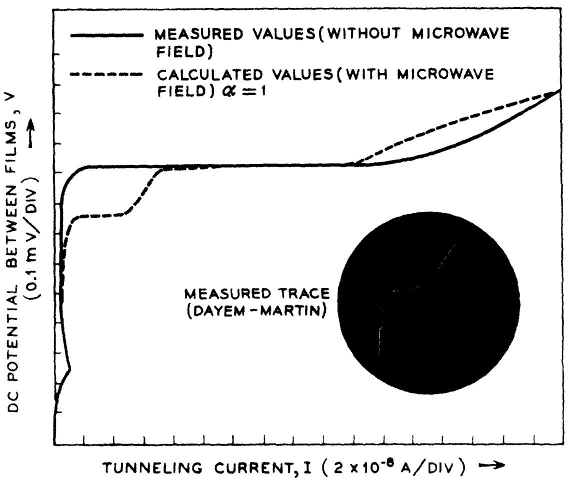

FIG. 2. Calculated bias voltage vs tunneling current of an Al-Al_2O_3-In diode for a microwave field of $\alpha=1$. For comparison, an oscillographic trace measured by Dayem and Martin is also shown below the computed curve. Microwave frequency $\nu=38.83$ kMc/sec; $\hbar\omega/e=0.16$ mV.

tunneling current becomes

$$I_{AB}'=c\sum_{n=-\infty}^{n=+\infty}J_n^2(\alpha)\int_{-\infty}^{+\infty}[f(E-eV_0)-f(E+n\hbar\omega)]\times\rho_A(E-eV_0)\rho_B(E+n\hbar\omega)dE. \quad (12)$$

A set of experimental data for the tunneling current without the microwave field [I_{AB} in (11)] was supplied by Dayem and Martin[6] and is reproduced in the solid traces of Figs. 2 and 3. From these data we may compute I_{AB}' (Eq. 12) for different α's. The calculated curves for I_{AB}' are shown as dashed traces in Figs. 2 and 3 for $\alpha=1$ and 2, respectively. These calculated curves for the tunneling current in the presence of the microwave field are in reasonably good agreement with the experimental results reported by Dayem and Martin.[1] However, the calculated electric field necessary to produce these results is far larger than that actually estimated in the experiment.

For example, consider the experiment at 24.2 kMc/sec. For $\alpha\cong1$, we require $V=10^{-4}$ V. For a dielectric layer 100 Å thick, this requires a field of 100 V/cm. One would, however, estimate that a field of only a few volts per centimeter was used in the experiments.

One further point is raised by Figs. 2 and 3. The calculated voltage-current traces show uniformly spaced steps along the voltage axis similar to those measured experimentally. Along the current axis, the steps extend roughly according to the Bessel functions $J_0^2(\alpha)$, $J_1^2(\alpha)$, $J_2^2(\alpha)\cdots$, since the density of states is sharply peaked at the edges of the energy gap. We thus see that in Fig. 3, one of the steps is almost missing on account of the fact that $J_0^2(\alpha)\cong0$ at $\alpha=2$. In the next case we find that those steps are distributed according to the integral of the Bessel functions and the steps will then be more uniform in the negative resistance region along the current axis.

IV. EFFECT OF MICROWAVE FIELD PARALLEL TO THE FILM SURFACE (CASE II)

In this case, we assume an rf electric field parallel to the film surface. The films are thin enough that the field penetrates through the diode despite the Meissner effect. The usual interaction Hamiltonian between the field and electrons is

$$H_I=-(ie\hbar/mc)(e^{i\omega t}+e^{-i\omega t})\mathbf{A}(\mathbf{r})\cdot\nabla+(e^2/2mc^2)(e^{i\omega t}+e^{-i\omega t})^2A^2(\mathbf{r}). \quad (13)$$

For convenience, Gaussian units are used throughout this paper unless otherwise specified. Here we have followed Mattis and Bardeen[10] and have taken the gauge $\nabla\cdot\mathbf{A}=0$, so that $\mathbf{E}=-(1/c)(\partial\mathbf{A}/\partial t)$. In order to estimate the field amplitude necessary to produce the desired effect, we may neglect the second term in (13) which is usually very samll. It is convenient to expand the vector potential, $\mathbf{A}(\mathbf{r})$, into Fourier components across the thickness of the film. Consider first film A, we have

$$\mathbf{A}(\mathbf{r})=(2\pi)^{3/2}\{\mathbf{A}_0+\sum_{q\neq0}\mathbf{A}(q)e^{i\mathbf{q}\cdot\mathbf{r}}\}, \quad (14)$$

where $\mathbf{q}$ is normal to the film surface, and $(\mathbf{A}(\mathbf{q})\cdot\mathbf{q})=0$. In addition,

$$|q|=\pi/l,\quad 2\pi/l\cdots\infty,$$

and l is the thickness of the film. Here we have separated A_0 from the rest of A's. A_0 is the component of the vector potential which is uniform over the film and the summation contains all the terms with $q\neq0$. As is shown later in this section, A_0 in film A is measured taking A_0 in film B as the reference, and the important quantity

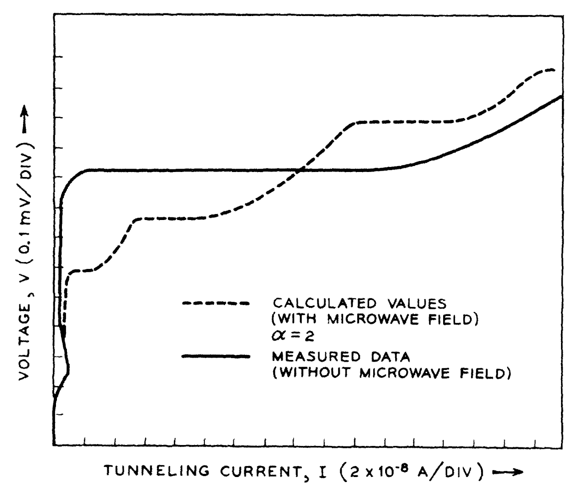

FIG. 3. Calculated bias voltage vs tunneling current of an Al-Al_2O_3-In diode for a microwave field of $\alpha=2$. Microwave frequency $\nu=38.83$ kMc/sec; $\hbar\omega/e=0.16$ mV.

[10] D. C. Mattis and J. Bardeen, Phys. Rev. **111**, 412 (1958).

is the difference of the A_0's in the two films. Let the Hamiltonian without the microwave field be H_0. With the field, the total Hamiltonian becomes

$$H = H_0 - \frac{ie\hbar}{mc}(e^{i\omega t}+e^{-i\omega t})(2\pi)^{3/2}\sum_{q\neq 0}\mathbf{A}(\mathbf{q})e^{i\mathbf{q}\cdot\mathbf{r}}\cdot\boldsymbol{\nabla} - \frac{ie\hbar}{mc}(e^{i\omega t}+e^{-i\omega t})(2\pi)^{3/2}\mathbf{A}_0\cdot\boldsymbol{\nabla}. \quad (15)$$

We denote the first two terms on the right-hand side of (15) by H_α, and the third term by H_β.

First, we solve H_α. The effect of those A's with $q\neq 0$ is known to cause the paramagnetic part of the conduction current, and the wave functions satisfying H_α are precisely those obtained by Mattis and Bardeen in the calculation of anomalous skin effect. For simplicity, we use the wave function for the normal conductor for illustration, it is

$$\psi(\mathbf{r},t) = e^{-iE_k t/\hbar}\Bigg[e^{i\mathbf{k}\cdot\mathbf{r}} - \sum_{q\neq 0} e^{i(\mathbf{k}+\mathbf{q})\cdot\mathbf{r}}(2\pi)^{3/2}\frac{e\hbar}{mc}\mathbf{A}(\mathbf{q})\cdot\mathbf{k} + \left(\frac{e^{i\omega t}}{E_{k+q}-E_k+\hbar\omega}+\frac{e^{-i\omega t}}{E_{k+q}-E_k-\hbar\omega}\right)\Bigg]. \quad (16)$$

Here the plane wave approximation is used, $e^{i\mathbf{k}\cdot\mathbf{r}}$ is the wave function satisfying H_0 of an electron having energy E_k and momentum $\hbar\mathbf{k}$. The terms containing A's are obtained from first-order perturbation theory. We have applied the gauge $\boldsymbol{\nabla}\cdot\mathbf{A}=0$ so that

$$\mathbf{q}\cdot\mathbf{A}=0. \quad (17)$$

For the superconductor, we need the matrix elements given in the Bardeen-Cooper-Schrieffer (BCS)[11] theory. The resultant wave function has the same general appearance.

Next we use (16) as (2) in Sec. II, and multiply it by a factor

$$\sum_{n=-\infty}^{n=+\infty} B_n e^{-in\omega t}$$

as in (4). We then substitute the product into the total Hamiltonian (15) and follow exactly the procedures shown in (5), (6), and (7). We obtain finally, after applying the relation (17), the wave function which satisfies the total Hamiltonian (15) as

$$\psi_k(r,t) = e^{-iE_k t/\hbar}\sum_{n=-\infty}^{n=+\infty} J_n(\alpha) \times e^{-in\omega t}\Bigg[e^{i\mathbf{k}\cdot\mathbf{r}} - \sum_{q\neq 0} e^{i(\mathbf{k}+\mathbf{q})\cdot\mathbf{r}}(2\pi)^{3/2}\frac{e\hbar}{mc}\mathbf{A}_q\cdot\mathbf{k} \times\left(\frac{e^{i\omega t}}{E_{k+q}-E_k+\hbar\omega}+\frac{e^{-i\omega t}}{E_{k+q}-E_k-\hbar\omega}\right)\Bigg], \quad (18)$$

where

$$\alpha = (2\pi)^{3/2}(2e\mathbf{A}_0\cdot\mathbf{k}/\omega mc). \quad (19)$$

[11] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).

To calculate the tunneling current, we may neglect as the first-order approximation, the terms involving the A_q's with $q\neq 0$. Equation (18) is now reduced to (8), and the density of electrons having energy $(E_k+n\hbar\omega)$ is again proportional to $J_n^2(\alpha)$. Caution must be exercised now since α is dependent on the momentum of the electron as shown in (19), and proper integration in momentum space must be made to calculate the total tunneling current.

Of importance to this calculation is the fact that α increases with the component of $\mathbf{k}$ parallel to $\mathbf{A}$, whereas the tunneling probability increases with the component of $\mathbf{k}$ normal to $\mathbf{A}$. We also notice that when we expand the electronic wave functions in films A and B in the form shown in (18), the effect of the microwave field on the tunneling current is dependent on $(\alpha_A-\alpha_B)$ (i.e., on the difference between the A_0's in the two films) not α_A or α_B alone. A detailed calculation has been carried out for this case. To obtain the same effect as for $\alpha=1$ in case I, we need a difference of transverse electric fields in the two films of the order of 4 V/cm. The estimated value in the experiment is many orders smaller.

V. THE DIODE AS A STRIP-LINE STRUCTURE

The thin-film diode is a sort of strip-line structure in which waves may propagate. The superconducting strip line has been analyzed by Swihart.[12] It is interesting to calculate the field distribution in such a structure and evaluate its effects.

We assume that the films and the aluminum oxide layer between them are each 100 Å thick. London's penetration depth is about 500 Å. With the mean free path limited by the thickness of the film, and taking a coherent distance $\xi_0=2500$ Å, we have a penetration depth for the films considered,

$$\lambda = 500\ \text{Å}(2500/100)^{1/2} = 2500\ \text{Å}.$$

We assume that both films have the same penetration depth.

In the transmission mode of a strip line there are E_x, H_y, and E_z fields. Here x is the direction normal to the films and z is the direction of propagation. It has been shown[12] that the ratio of the velocity of light in free-space to the propagating velocity of the structure is

$$\frac{c}{v} = \left(1+\frac{\lambda_A}{d}\coth\frac{\lambda_A}{l_A}+\frac{\lambda_B}{d}\coth\frac{\lambda_B}{B}\right)^{1/2} \cong 104. \quad (20)$$

Also we have in the insulating region

$$|E_z/H_y| = 2\pi(\lambda_A^2/\lambda_0 l_A) \cong 3.17\times 10^{-3}, \quad (21)$$

and in the two films

$$|E_x/H_y| = (c/v)(1/\epsilon_2) = 12, \quad (22)$$

[12] J. C. Swihart, J. Appl. Phys. **32**, 105 (1961).

where λ_A, λ_B and l_A, l_B are the penetration depths and the thicknesses of the films A and B, respectively. d is the thickness of the aluminum oxide layer which is assumed to have a dielectric constant, $\epsilon_2=8.6$, λ_0 is the free-space wavelength and in Gaussian units the impedance of the free-space is unity. We notice that for very thin films, E_z is uniform in the films and E_x and H_y are uniform in the insulating region.

We see here from (21) and (22) that

$$|E_z/E_x|\cong 2.64\times 10^{-4}. \tag{23}$$

In the transmission line mode, the E_z fields in the two films are in the opposite directions. From the calculation made in Secs. III and IV, we may conclude that E_z field of 2 V/cm should produce the same effect as a E_x field of 100 V/cm. Since the ratio of E_z/E_x in (23) is much smaller, it is clear that E_x field in the strip line must produce the dominant effect. The film surface used in the experiments is about 1 mm^2, which according to (20), may contain 10 wavelengths along each side. Suppose now that the transmission line mode is excited by the E_x field of the cavity. One would estimate that a Q of the strip-line structure of more than 6000 is necessary in order to produce the observed effect. Such a high Q should cause some resonant response due to the diode alone and this was not observed in any of the experiments.

VI. DISCUSSION

We have been able to account qualitatively for the multiphoton-assisted tunneling current in superconducting diodes. The calculations are based on the assumption that the electronic energy levels in the diode are modulated adiabatically by the presence of a microfield whose photon energy is smaller than the energy gap. There remains a marked quantitative disagreement between theory and experiment, however; the effect occurs for field strengths at least an order of magnitude smaller than would be expected from the theory.

No nonlinear effects in the conduction current are to be expected from these small fields; and in fact it may easily be shown that the wave functions (8) and (18) give precisely the same paramagnetic and diamagnetic parts of the conduction current as those calculated by Mattis and Bardeen. The effect of the field is rather to alter the effective density of states versus energy, and this change shows up directly in the tunneling current.

The parameter important to this calculation is the ratio of the change in energy of the electrons, as a result of the microwave field, to the photon energy. This ratio is denoted as α. The calculated results, as shown in Figs. 2 and 3, agree well with the experimental results of Dayem and Martin. However, values of α near unity are required for quantitative agreement, and the microwave fields used in the experiment were considerably smaller than this would necessitate. Despite the lack of quantitative agreement, we feel that the adiabatic assumption is probably correct.

Note added in proof. It has been called to the authors' attention that a similar expression for the effect for a normal electric field as described here in case I, has been obtained by Cohen, Falicov, and Phillips.[13] In addition, they have given the result of the modulation of the energy gap by a magnetic microwave field. which yields

$$\alpha=E_{\text{gap}}H_{\text{rf}}/\hbar\omega H_c,$$

where E_{gap} is the energy of the gap, H_{rf} is the microwave magnetic field, and H_c is the critical field of the superconducting film. Let us consider an aluminum thin film. For $E_{\text{gap}}\cong 0.32\times 10^{-3}$ eV, $H_{\text{rf}}=1$ Oe, $H_c\cong 6000$ Oe, and $\hbar\omega=10^{-4}$ eV, α is in the order of 5×10^{-4}. A magnetic field of 1 Oe would produce a maximum electric field about 300 V/cm in a microwave cavity, and α would be 3 in the case of normal electric field as considered in this paper.

[13] M. H. Cohen, L. M. Falicov, and J. C. Phillips, *Proceedings of the Eighth Conference in Low-Temperature Physics, 1962* (to be published).

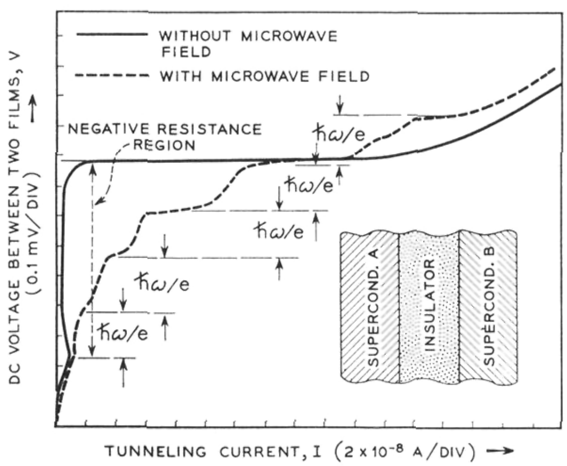

FIG. 1. Bias voltage vs tunneling current of a superconducting Al-Al_2O_3-In diode as measured by Dayem and Martin with and without the microwave field. $\hbar\omega/e = 0.16$ mV.

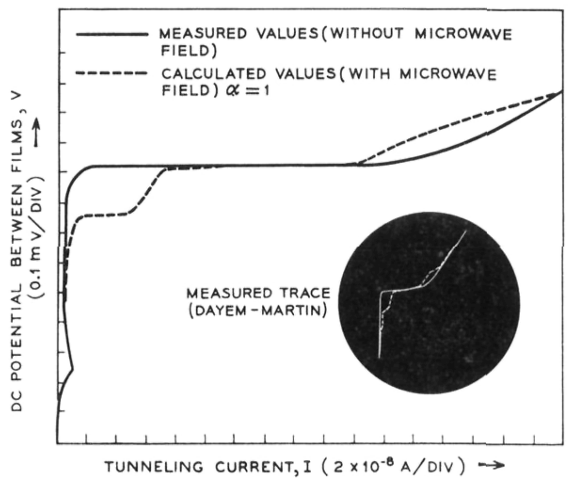

FIG. 2. Calculated bias voltage vs tunneling current of an Al-Al_2O_3-In diode for a microwave field of $\alpha=1$. For comparison, an oscillographic trace measured by Dayem and Martin is also shown below the computed curve. Microwave frequency $\nu=38.83$ kMc/sec; $\hbar\omega/e=0.16$ mV.